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Single Molecule Source Reagents for CVD of Beta Silicon Carbide

Monthly Report

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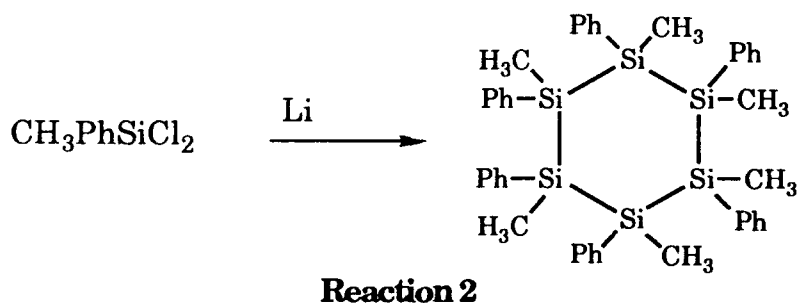
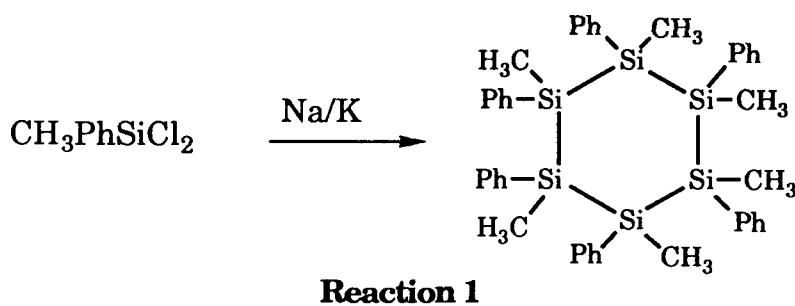


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Beta silicon carbide is an excellent candidate semiconductor material for demanding applications in high power and high temperature electronic devices due to its high breakdown voltage, relatively large band gap, high thermal conductivity and high melting point. Use of silicon carbide thin films is hampered, however, by the inability to reproducibly grow stoichiometric films free from excess silicon or carbon. The principal difficulty is that absolutely reproducible flows of the source gases cannot be provided with existing gas flow control equipment. The very finest gas flow control equipment has been used in the CVD systems, but small disparities remain between successive deposited films. The production of practical beta silicon carbide devices has been hindered by the lack of a reproducible process for deposition of stoichiometric beta silicon carbide films.

The goal of the Phase I effort is to synthesize single molecule source reagents, study their decomposition pathways, and investigate their use for the CVD of beta silicon carbide. The ultimate objective is to produce a high purity, unimolecular source reagent which will introduce equivalent amounts of *reactive* silicon and carbon to the growth surface and lead to growth of intrinsic, stoichiometric β -SiC for device applications.

During the month of August numerous attempts were made to isolate crystalline solids from the crude products of the two reactions shown below:



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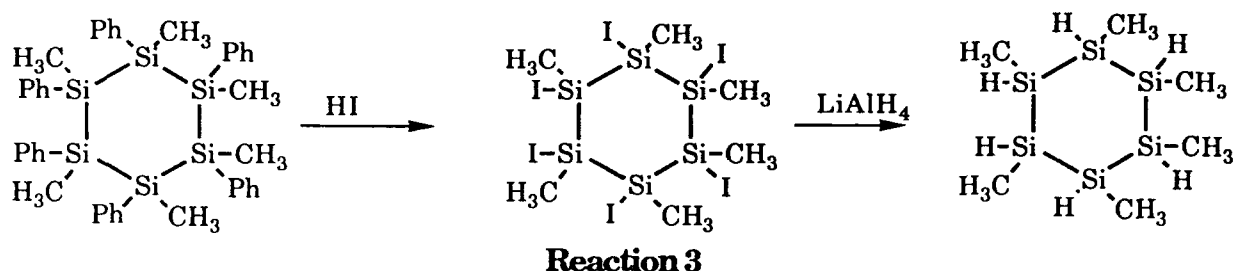
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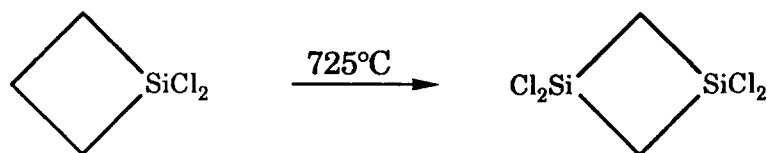
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These efforts were not successful and instead the crude product from the Na/K was allowed to react with HI and the resulting material reduced with lithium aluminum hydride (Reaction 3)



This procedure did not result in isolation of the desired product. Due to the low yield in the initial step (reported yield: 10%) synthesis of a reasonable quantity of the desired product appears unlikely.

Further work was also done on the synthesis of 1,4-disilacyclobutane. The pyrolysis procedure (reaction 4) which is the first step in the synthesis is painstakingly slow, allowing isolation of only a few grams of material each day. However, semi-continuous operation has allowed isolation of ~30grams of this material. A small quantity has been reduced (reaction 5) to produce the desired 1,4-disilacyclobutane. This material contained two impurities. One of these appears to arise from ring-opening which is most likely occurring during the reduction step. This should be preventable by evaporating the product from the reaction mixture as it is formed.



Experimental details are given below:

Hexamethylcyclohexasilane

Acetone was removed from the viscous oil which had resulted from the reaction of PhMeSiCl_2 with Na/K alloy. A few ml of benzene was used to transfer the material to a Schlenk tube via cannula. HI was bubbled into the benzene solution which caused it to go from colorless to orange-brown. The solution was stirred under HI for several days, introducing fresh HI periodically. HI and benzene was stripped in vacuo, yielding a red-brown oil. An excess of LAH (based upon the original quantity of PhMeSiCl_2) was added to 20 ml of tetraglyme and to this was added the red-brown oil which had been dissolved in 20 mL of tetraglyme (stirring the red-brown oil overnight in tetraglyme had resulted in the

dissappearance of the red color). Substantial frothing (gas evolution) was observed throughout the addition which was accomplished over a few hrs. The suspension was stirred overnight, and then a trap was set up to collect the volatile components in the flask. Several evacuate-equilibrate cycles were performed including some warming of the reaction vessel to promote transfer. A drop of material was observed in the trap which evaporated on warming to room temperature and is therefore clearly too volatile to be the desired product.

1,4-disilacyclobutane

Sublimed tetrachlorodisilacyclobutane (3.16g 0.014 moles) was added to a small addition funnel and an additional portion (10.0 g, 0.044 moles) was added to a 125 mL Wheaton bottle in the drybox. Lithium aluminum hydride (2.6 g, 0.068 moles) was loaded into a solids addition tube. This and the addition funnel were attached to a 500 mL 3-neck r.b. flask and removed to the hood. 25 mL of tetraglyme was used to dissolve the tetrachlorodisilacyclobutane in the funnel and another 100 mL of 4-glyme used to dissolve the 10 g. 100 mL of 4-glyme was added to the reaction flask which was then cooled to -5°C in an ice/water bath. The LiAlH_4 was added to the cold 4-glyme in small portions. Meanwhile, a trap was attached to the system, evacuated and dried with a heat gun. The trap was cooled to -78°C and the system was vented through this trap. The solutions of tetrachlorodisilacyclobutane were added dropwise via the addition funnel over 30 min. The reaction mixture was then stirred for an additional hour at room temperature. The trap was then cooled to -196°C and the volatiles were transferred by several evacuate-equilibrate cycles. A power failure caused the transfer to be completed the following day. The volatiles were then transferred from the trap to a Kontes ampule. Yield: 1.9 g, 0.021 moles, 37%. GC/MS of the resulting colorless liquid revealed two significant impurities. The impurity which is more volatile than the product is likely a ring-opened species, while the one of higher volatility appears to be $\text{H}_2\text{Si}(\text{CH}_2)_2\text{SiHMe}$.